INFLUENCE OF Ag-DOPED ON MAGNETIC AND ELECTRICAL TRANSPORT PROPERTIES IN $Pr_{0.75}Na_{0.25}MnO_3$ MANGANITES

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ABSTRACT

The manganites compound in which composed of the manganese oxide are attract considerable attention among researcher due to their electrical and magnetic properties at lower temperature. However, most of the doping does not totally induce the ferromagnetic-metallic (FMM) state probably due to dopant level. As such, the influence of ion doped on host composition need to be further investigated. In this study, the monovalent doped $Pr_{0.75}Na_{0.25-y}Ag_yMnO_3$ (y = 0 – 0.10) manganites was prepared by solid-state reaction method. All samples have been characterized using the X-ray diffraction (XRD) and scanning electron microscope (SEM) as well as DC electrical resistivity and AC susceptibility measurement to clarify the influence of Ag-doped on monovalent doped manganites. XRD analysis revealed all samples consists of essentially single phase and crystallized in an orthorhombic structure with space group *Pnma*. SEM images of $Pr_{0.75}Na_{0.25-y}Ag_yMnO_3$ compound shows the successful substitution of Ag^+ ions with the enhancement of the grains boundaries and sizes as well as the compaction of particles. On the other hand, susceptibility and resistivity measurements showed that the y = 0 sample exhibits anti-ferromagnetic insulating behaviour with Néel temperature, T_N ~ 125 K. Interestingly, the FMM transition was observed for y = 0.05 with the metal insulator transition temperature, T_{MI} ~ 110 K and Curie temperature, T_C ~ 123 K. However, increasing of Ag-doped up to y = 0.10 showed an insulating behaviour and paramagnetic-ferromagnetic transition with T_C around 126 K. Apart from that, the resistivity behaviour at temperature region above T_{MI} for y = 0 – 0.10 was found to fit well with the variable range hopping model (VRH) with the increasing of hopping and activation energy as the Ag concentration increased. While for the metallic region which is below the T_{MI}, the resistivity data of y = 0.05 was fitted well with the combination of domain/grain boundary, electron-electron and electron-magnon scattering.

ABSTRAK

Kompaun *manganites* yang terdiri daripada oksida mangan telah menarik perhatian penyelidik kerana sifat elektrik dan magnetnya pada suhu yang lebih rendah. Walau bagaimanapun, kebanyakan doping tidak sepenuhnya menginduksi keadaan feromagnetik-logam (FMM) mungkin disebabkan tahap dopan. Oleh yang demikian, pengaruh ion yang dikenakan terhadap komposisi bahan utama perlu disiasat selanjutnya. Dalam kajian ini, monovalen $Pr_{0.75}Na_{0.25-y}Ag_{y}MnO_3$ (y = 0 – 0.10) *manganites compound* telah disediakan dengan menggunakan tindakbalas keadaan pepejal. Semua sampel telah dikaji dengan menggunakan *X-ray diffraction* (XRD) dan *scanning electron microscope* (SEM) serta *DC electrical resistivity* dan *AC* susceptibility untuk menjelaskan pengaruh Ag *doped* terhadap monovalen *manganites*. Analisis XRD mendedahkan semua sampel terdiri pada dasarnya fasa tunggal dan menghablur dalam satu struktur ortorombus dengan kumpulan ruang *Pnma*. Kajian dengan mengunakan SEM telah menunjukkan keberkesanan penambahan Ag ions terhadap $Pr_{0.75}Na_{0.25-y}Ag_{y}MnO_3$ dengan peningkatan *grain boundary* dan size grain serta pemadatan zarah. Sebaliknya, pengukuran *DC electrical resistivity* menunjukkan bahawa sampel y = 0 mempamerkan tingkah laku menebat dan anti feromagnet, T_N ~ 125 K. Menariknya, peralihan logam-feromagnet telah diperhatikan untuk sample y = 0.05 dengan nilai T_{MI} ~ 110 K yang mana peralihan ini disebabkan oleh pemulihan *double exchange (DE) mechanism* hasil daripda kelemahan *Jahn Teller (JT) effect*. Walau bagaimanapun, peningkatan Ag-doped sehingga y = 0.10 telah telah menyebabkan sampal bersifat penebat dan berlaku peralihan paramagnetic-ferromagnetik dengan peningkatan nilai T_C ~ 126 K. Selain itu, *resistivity behaviour* pada suhu tinggi, atas T_{MI} for y = 0 – 0.10 didapati sepadan dengan menggunakan *variable range hopping* (VRH) yang mana *activation* dan *hopping energy* meningkat apabila Ag-doped meningkat disebabkan oleh herotan MnO_6 oktahedron. Untuk metallic region, suhu dibawah nilai T_{MI}, sifat kerintangan untuk y = 0.05 sepadan dengan gabungan *domain / grain boundary, electron-electron* dan *electron-magnon scattering*.

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LIST OF SYMBOLS AND ABBREVIATIONS

AFM	-	Antiferromagnetic
Ba	-	Barium
B	-	Resistivity coefficient
Ca	-	Calcium
CO	-	Charge-orbital ordering
CrO_3	-	Chromium oxide
Cr	-	Chromium
CMR	-	Colossal magnetoresistance
DE	-	Double exchange
Dy	-	Dysprosium
D	-	Density
E_a	-	Activation energy
E_h	-	Hopping energy
FM	-	Ferromagnetic
I	-	Current
JT	-	Jahn Teller
K	-	Kelvin
k_B	-	Boltzmann’s constant
La	-	Lanthanum
Mn	-	Manganese
MnO_2	-	Manganese oxide
Na	-	Sodium
Na_2CO_3	-	Sodium carbonate
Nd	-	Neodymium
$N(E_F)$	-	Density of states
P	-	Resistivity
ρ_{0m}	-	Residual resistivity

PM	-	Paramagnetic
Pr	-	Praseodymium
Pr_2O_3	-	Praseodymium oxide
R_h	-	Mean hopping distance
S	-	Needle spacing
Sr	-	Strontium
T	-	Temperature
T_C	-	Curie temperature
T_{MI}	-	Transition temperature (metal - insulator)
T_N	-	Néel temperature
T_{0m}	-	Characteristic temperature
V	-	Volume
XRD	-	X-ray diffraction measurement

LIST OF APPENDICES

LIST OF PUBLICATIONS

1. Khairulzaman, N., Ibrahim, N., & Shamsuddin, S. (2018). Impact of Ag Doped on the Ferromagnetic-Metallic Transition in $Pr_{0.75}Na_{0.25}MnO_3$ Manganites. International Journal of Engineering & Technology, 7(4.30)68-71
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2. Zawawi, R. A., Khairulzaman, N., Shamsuddin, S., & Ibrahim, N. (2018). Comparative Study on Structural, Electrical Transport and Magnetic Properties of Cr-Doped in Charge-Ordered $Pr_{0.75}Na_{0.25}Mn_{1-x}Cr_xO_3$ and $Nd_{0.75}Na_{0.25}Mn_{1-y}Cr_yo_3$ Manganites. International Journal of Engineering & Technology, 7(4.30)75-79
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3. Zawawi, R. A., Khairulzaman, N., & Shamsuddin, S. (2017). Effect of Cr-doped on Crystalline Phase, Surface Morphology and Electrical Properties of Charge-Ordered $Nd_{0.75}Na_{0.25}MnO_3$ Ceramics. Journal of Science & Technology, Vol. 9 No.3 p. 45-48.

CHAPTER 1

INTRODUCTION

1.1 Background of study

In recent years, discovery of the properties of magnetic material among the manganite compound which is composed of the manganese with the general formula $Re_{1-x}A_xMnO_3$ where Re is trivalent rare-earth ion (Pr^{3+}, La^{3+}, Nd^{3+}, Dy^{3+}) and A is a divalent (Ca^{2+}, Sr^{2+}, Cr^{2+}, Ba^{2+}) or monovalent (Ag^{+}, K^{+}, Na^{+}) alkaline earth ion have received remarkable attention due to the discovery of Colossal magnetoresistance (CMR) effect. The great attentions were increased due to their unique properties such as charge-ordering (CO), ferromagnetic-metallic (FMM) transition as well as have the high potential application at lower temperature (Sankarrajan *et al*., 2009; Orlova *et al*., 2013). The investigation on the rare earth manganite become more popular and interesting when it was related with the CO which is known as the ordering of the metal ions in different oxidation states. Generally the CO was localized the charge and restrict the electron to hope from one site to another site and causes the manganite exhibit the insulating or semiconducting behaviour (Raveau *et al*., 2000; Liu *et al*., 2007).

In previous studies, the CO state in the divalent doped manganite such as $Nd_{0.5}Ca_{0.5}MnO_3$ (Liu *et al*., 2007), $Pr_{0.5}Sr_{0.5}MnO_3$ (Pramanik, 2013) and $Pr_{0.67}Sr_{0.33}MnO_3$ (Hcini *et al*., 2011) were often observed compared to monovalent doped manganite such as $Nd_{0.75}Na_{0.25}MnO_3$ (Li *et al*., 2004) and $Pr_{0.75}Na_{0.25}MnO_3$ (Jir'ak *et al*., 2002). There are two different oxidation states exist in manganese ion for the hole-doped manganite which are Mn^{3+} and Mn^{4+} (Jirak *et al*., 2000) where the ratio of Mn^{3+} and Mn^{4+} among the CO system for the half-doped of $RE_{0.5}A_{0.5}MnO_3$ is

1:1 and it has received fair attention due to their unique magnetic and transport properties (Fan *et al*., 2008; Yadav *et al*., 2012). The transformations of manganite behaviour commonly were suggested due to the double exchange (DE) mechanism and Jahn Teller (JT) effect. DE mechanism involved the movement of electron in between Mn^{3+}, oxygen O^{2-} and Mn^{4+} that favour to the ferromagnetic metallic (FMM) state (Gor'kov & Kresin, 2004) while Jahn Teller effect known as a dominant of Mn^{3+} ion causes the behaviour of paramagnetic insulating (Millis *et al*., 1995).

Furthermore, the physical properties of manganite can be affected by the change of tolerance factor (г), average size of A site cation and mismatch effect factor (σ^2) of manganite (Shaikh & Varshney, 2014). Several reports suggested that the substitution of transition-metal element at A-site of manganite were influence the electro-magnetic properties due to the dislocation of e_g electron as a result of random distribution of A-site cations (Shamsuddin *et al.*, 2013a; Tang *et al.*, 2008a). Previous study also shows that the increment of Ag^+ on $Pr_{0.5}Sr_{0.5-x}Ag_xMnO_3$ and $La_{0.67}Pb_{0.33}MnO_3$ was induced the FMM phase with the change of Curie temperature (T_C) and metal-insulator transition temperature (T_{MI}) (Mtiraoui *et al.*, 2011; Tarhouni *et al.*, 2017). Meanwhile, the substitution of Na^+ by K^+ in $Pr_{0.75}Na_{0.25-x}K_xMnO_3$ was reported to induce the FMM phase with the increasing of T_C and T_{MI} suggestively due to the K^+ ion is larger than Na^+ ion which causes an increasing in tolerance factor and e_g electron transfer, which promoted the DE mechanism (Rozilah *et al.*, 2017). This observed behaviour indicated the important role in the suitable substitution of some element at A–site can affect the magnetic and electrical transport properties. Unfortunately, a lot of holes doped manganite investigations were reported in the charge ordered divalent doped manganite compared to the charge ordered monovalent doped manganite. Therefore, in this work the investigation monovalent holes doped manganite is requiring a further investigation especially in the magnetic and electrical transport properties.

Meanwhile, scanning electron microscope (SEM) measurement have been useful in providing valuable information on the surface morphology of the compound including manganites (Hcini *et al.*, 2011; Tourhoni *et al.*, 2017). For instance, the substitution of K^+ ions on $Nd_{0.75}Na_{0.25-x}K_xMnO_3$ were enhanced the grain boundary and grain size as the K^+ content increased due to the different ionic radius between K^+ ions and Na^+ ions (Razali *et al.*, 2018). In addition, the substitution of Cr^{3+} and

Co^{3+} at Mn site of the $Pr_{0.75}Na_{0.25}MnO_3$ were also change the grain boundary and grain size as well as the porosity of the samples due to the difference of ionic radius (Zawawi *et al.,* 2017; Ab Mannan *et al.,* 2017). Study on $Pr_{0.75}Na_{0.25-y}Ag_yMnO_3$ is expected to provide more information about manganites compound as well as deliver new information on the morphological study.

On the other hand, studies of electrical transport analysis in the manganite have been useful in order to understand the variation of resistivity behaviour in manganite compound. Based on the previous study, the electrical properties can be explained by applying the scattering model for the metallic region and small polaron hoping (SPH) with variable range hoping (VRH) model for insulating region (Tozri *et al.,* 2014; Munirathinam *et al.,* 2012; Ghani *et al.*, 2012). Previous study claimed that the manganite compound of $Bi_{0.3}Pr_{0.3}Ca_{0.4}Mn_{1-x}Cr_xO_3$ was exhibit the metal-insulator (MI) transition. The temperature resistivity independent graph for the metallic region was found to be fit well with the electron-electron scattering, the Kondo like effect and electron–phonon interaction. While the VRH and SPH models were used to explain the resistivity in the insulating region with the decreasing of hopping energy, E_h and activation energy, E_a as the Cr^{3+} increased (Asmira *et al.*, 2018). This phenomenon was suggested due to the increase in delocalization of charge carriers attributed to the weakening of Jahn-Teller effect as a result of decrease in Mn^{3+}. The substitutions hence lead to decreased in MnO_6 octahedral distortion (Asmira *et al.*, 2018; Rozilah *et al.*, 2017). In this study, the element transition of silver with single ion have been substituted at the A-site of the $Pr_{0.75}Na_{0.25-y}Ag_yMnO_3$ in order to investigate the influences of Ag^+ ions on structure, magnetic and electrical transport properties as well as determining the variation of resistivity behaviour of manganite compound.

1.2 Problem statement

Research on monovalent-doped manganites $Pr_{0.75}Na_{0.25}MnO_3$ has tremendously capture researchers' attention among the rare-earth based manganites (Kozlenko *et al.*, 2004; Zhang *et al.*, 2005) due to the existence of charge ordering (CO) between Mn^{3+}/Mn^{4+} with the ratio of 1:1 and the CO transition was observed at high temperature compared to anti-ferromagnetic (AFM) transition (T_{CO} ~ 215 K, T_N ~ 175 K) (Jirak *et al.*, 2002; Hejtmanek *et al*., 2001; Satoh *et al*., 2002). The investigation on this compound become interesting with the doping of small amount of element at A/Mn-sites in which it can influences the origin of the compound including the CO state (Elyana *et al.*, 2018; Zawawi *et al.*, 2018; Li *et al.*, 2007). Previous study on monovalent doped manganites $Pr_{0.75}Na_{0.25-x}K_xMnO_3$ shows that the small level doping of potassium, K^+ (x = 0.05 0.10) was exhibit AFM and the increasing of K^+ doped into x = 0.15 – 0.20 was induced the ferromagnetic-paramagnetic behaviour. Then, for the electrical behaviour, the increasing of K^+ doping with x = 0.05 – 0.20 was enhanced the metal-insulator transition due to enhancement of double exchange (DE) mechanism as a result of large K^+ ions doped (Rozilah *et al*., 2017). While, the study on $Pr_{0.75}Na_{0.25}Mn_{1-x}Ru_xO_3$ ($0.01 \le x \le 0.10$) shows that the increment of Ru content was shifted the T_C and T_{MI} to higher temperatures due to the enhancement of double exchange mechanism (Elyana *et al*., 2018). It is clear in the literature that the substitution of some element in manganites compound plays an important role which can enhance the properties of manganite such as magnetic and electrical transport properties. However, most of the doping does not totally induce the FMM state probably due to dopant level. As such, the influence of ion doped on host composition need to be further investigated especially on the substitution at A–site of manganites in order to understand the role of ions doped in the manganites compound. Hence, study on the $Pr_{0.75}Na_{0.25-y}Ag_yMnO_3$ can provide a significant information mainly on the surface morphology, electrical and magnetic properties and to our knowledge, the $Pr_{0.75}Na_{0.25-y}Ag_yMnO_3$ system has not been previously reported. Moreover, a few study stated that the Ag^+ ion doped in manganites compound can generate the FMM transition and enhance the curie temperature (Tc) as well as metallic insulator transition temperature (T_{MI}) (Yin *et al*., 2015; Battabyal & Dey, 2006; Thaljaoui *et al*., 2017). On the other hand, this study also cover the analysis of the experimental electrical resistivity data where it provide give a better understanding on the electrical behaviour of $Pr_{0.75}Na_{0.25-y}Ag_yMnO_3$ by applying the electron scattering and hopping process.

1.3 Objective of study

i. To determine the crystalline structure of $Pr_{0.75}Na_{0.25-y}Ag_yMnO_3$ using powder X-ray diffraction (XRD) measurement as well as determining the surface morphology using the scanning electron microscope (SEM).
ii. To elucidate the effect of Ag-doped at A-site in $Pr_{0.75}Na_{0.25}MnO_3$ by determining the magnetic and electrical transport properties using AC susceptibility and DC electrical resistivity measurement.
iii. To analyze the electrical resistivity data using scattering and polaron hoping model and reporting the possible mechanism in the compound of $Pr_{0.75}Na_{0.25}MnO_3$ manganites with the Ag doping.

1.4 Scope of study

The magnetic material of charge ordered $Pr_{0.75}Na_{0.25-y}Ag_yMnO_3$ manganite with the composition of y = 0, 0.05 and 0.10 were investigated in order to elucidate the effect of Ag^+ ions on the structure, surface morphology, magnetic properties and electrical properties as well as determine the variation of resistivity behaviour. The bulk samples were synthesize using the solid-state reaction method involving several processing such as mixing, grounding, calcination, pelleting and sintering process. The confirmation of crystalline structure of bulk samples were carried out using the powder X-ray diffraction (XRD) measurement with Cu Kα radiation and the surface morphology of $Pr_{0.75}Na_{0.25-y}Ag_yMnO_3$ were determined using Hitachi SU1310 scanning electron microscope (SEM) with magnification 5 kX. Meanwhile, for the AC susceptibility (χ') of the samples was measured using an AC susceptometer system manufactured by CryoBIND-T system in conjunction with its real component resolved using a lock-in amplifier. Other than that, the temperature dependent resistivity (ρ) for all samples were measured using the standard four point-probe technique contact with silver paint where the sample was placed on the head of a closed cycle refrigerator device at low temperature in a range of 50 – 300 K. To further understand the resistivity behaviour of $Pr_{0.75}Na_{0.25-y}Ag_yMnO_3$ for the temperature below and above the T_{MI}, the resistivity data were analysed using the scattering and hoping model.

1.5 Significant of study

In past few decades, the investigation on the magnetic material especially on the manganites compound have received a remarkable attention due to their high potential application at lower temperature such as magnetic field sensor, electrical field effect devices and bolometric uncooled infrared (IR) sensor using the metal-insulator (MI) transition at the curie temperature. In this study, the investigation on the monovalent hole doped manganite $Pr_{0.75}Na_{0.25}MnO_3$ is important in ordered to elucidate the effect of Ag doping on the structure, surface morphology, magnetic properties and electrical transport properties. This study is expected to provide additional information and knowledge especially on the surface morphology and FMM transition because this study is still not clearly reported. Apart from that, this study have been providing significant information on the resistivity behaviour of monovalent hole doped manganite compound by applying the scattering and polaron hoping models.

CHAPTER 2

LITERATURE REVIEW

2.1 Introduction

This chapter explained about the information of relevant studies which related to this research work that elucidates the effect of Ag-doping on the structure, surface morphology, magnetic and electrical properties as well as analyse the electrical behaviour using polaron hopping model and scattering model. This chapter provided with some basic information and knowledge related with the research for the basic understanding especially related with the manganites compound for instance, structure of manganites, magnetic behaviour of manganites and interaction that involved in the manganites compound. Furthermore, this chapter also discussed the previous studies of manganites compound which focused on the effected of doping on the structure, surface morphology, magnetic and electrical transport properties as well as electrical behaviour analysis in the monovalent and divalent doped manganites.

2.2 Structure of manganites

2.2.1 Ceramic

Generally, solid materials have been grouped into three basic categories which are metals, ceramics and polymer based on their chemical makeup and atomic structure. In the metals group, the compound is composed by one or more metallic element (iron, aluminium, copper and titanium) and non-metallic elements (oxygen, carbon and nitrogen) while for the polymer grouped, many of them are organic compound that is chemically based on carbon, hydrogen and other nonmetallic elements. For the ceramics, the compounds are between the metallic and non-metallic elements where this material more frequently oxides, nitrides and carbides. For examples aluminium oxide, silicon dioxide, silicon carbide, silicon nitride and for this study, the manganites compound was grouped as a ceramic compound due to their atomic structure.

Ceramic is quite good material due to their mechanical behaviour where these materials are relatively stiff and strong stiffnesses and strengths as well as very hard. Earlier, ceramic materials have exhibit extreme brittleness and highly susceptible to fracture but nowadays with the technologies, ceramics materials are being engineered to improve resistance towards fracture. Other than that, these materials also have low electrical conductivity and have more resistant towards high temperature and harsh environments compared to metal and polymers. Some of the oxides ceramics tends to exhibit magnetic behaviour for example Fe_3O_4. For the structure properties, ceramic materials are composed by at least two elements and often more with the complex crystal structure compare with others, for example, AX-type, A_mX_p-type and $A_mB_nX_p$-type crystal structure. The crystal structure of these materials depends on their atomic bonding which many ceramic exhibits a combination of ionic and covalent bond type (Callister & Rethwisch, 2013; Askeland & Phule, 2006).

2.2.2 Crystal structure of ceramics

The crystal structure is constructed by the lattice and basis. This can be described through the infinite repetition of identical groups of atoms which the group is known as basis meanwhile lattice is a set of mathematical points where the basis is attached. Every basis of atoms is identical to every other structure and composition. There are seven crystal systems which fill in the three-dimension space with the 14 distinct arrangement of lattice point known as Bravais lattices as shown in Figure 2.1. There are three relatively simple crystal structure which are simple cubic (SC), face-centred cubic (FCC) and body-centred cubic (BCC). For the SC crystal structure, the atoms are located only at the corners of the cube while for the FCC crystal structure, atoms were located at each of the corners and the centres of all the cube faces. Then for the

BCC crystal structure, the atoms were located at the corners and the centre of the cube.

For the ceramic materials, the crystal structures are generally more complex with others because these materials are composed of at least two elements and often more. The atomic bonding in this material commonly exhibits a combination of two bonding which is ionic and covalent bond where the crystal structure of this material being composed of electrically charged ions instead of atoms. The crystal structure of ceramic can be affected with two characteristics of component ions in crystalline ceramic materials which are the magnitude of the electrical charge on each of the component ions and the relative sizes of the cations and anions. As a regard for the first characteristic, the crystal must be electrically neutral which means that all the cations charge must balance by having an equal numbers of the negative charge of anions.

Then for the second characteristic, the size of cation and anions can influence the crystal structure due to their different size or ionic radii where the sizes of cations are ordinarily smaller than anions. In the ceramic materials, there have three types of crystal structure including AX-type, A_mX_p-type and $A_mB_nX_p$-type crystal structure. Commonly, the ceramic materials with the equal numbers of cations and anions are often referred to as AX compound where A denotes the cations and X the anion. AX compounds have several crystals structure, for example, rock salt structure, cesium chloride structure and zinc blend structure. For the A_mX_p -type, the charge on the cation and anions are not the same where m and/or p $\neq$ 1 for instance is AX_2 where this commonly found in fluorite (CaF_2). While for the $A_mB_nX_p$-type, this chemical formula is possible for the ceramic compound to have more than one type cations represent by A and B for example Barium titanate ($BaTiO_3$). This material has a perovskite crystal structure and generally this material could be described as ABO_3. Manganites compound with the general formula $Re_{1-x}A_xMnO_3$ was indicated as a perovskite structure based on the XRD analysis (Callister & Rethwisch, 2013; Askeland & Phule, 2006).

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